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Developing stable fine–grain microstructures by large strain deformation

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THE ROYAL SOCIETY

Developing stable fine-grain microstructures by large strain deformation

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Methods of deforming metals to large strains are reviewed and the process of equal channel angular extrusion is analysed in detail. The development of microstructure during large strain deformation is discussed, and it is concluded that the main criterion for the formation of a sub-micron grain structure is the generation of a sufficiently large fraction (greater than 0.7) of high-angle grain boundary during the deformation process. For aluminium alloys, it is found that a low-temperature anneal is required to convert the deformed microstructure into an equiaxed grain structure. The material, microstructural and processing factors that influence the formation of such fine-grain microstructures are discussed, and the stability of these microstructures at elevated temperatures is considered.

Keywords: sub-micron grain; grains; deformation; large strain; grain boundary

1. Introduction

The grain size of a metal has a large effect on its properties, and refinement of the grain size has many technological benefits. For example, at low temperatures, a small grain size may increase the strength and toughness of the material, and, at high temperatures, fine-grained alloys may become superplastic. This paper discusses the application of novel deformation processing methods to the production of sub-micron grain structures that are considerably smaller than those produced by conventional processing.

There are a number of methods of producing metallic materials with sub-micron grain sizes, including rapid solidification, powder metallurgy, and vapour condensation methods, and the production and properties of *nanostructured materials* is now a well-established field of materials science. However, most of these methods are only applicable to the production of very small quantities of material, often of unusual compositions, whereas the emphasis in this paper is on the production of fine-grain structures by methods that are based on the deformation of bulk metal, and that are applicable to larger quantities of conventional structural alloys.

The as-cast grain size of most industrial alloys is generally large (greater than 100 µm), and further grain refinement is achieved by thermomechanical processing. In alloys that undergo massive solid-state phase transformations, such as steels and titanium alloys, grain refinement may be obtained via such transformations, and for steels, controlled rolling during the phase transformation ($\gamma \rightarrow \alpha$) may result in ferrite grain sizes of less than 5 µm. For aluminium alloys, fine-grain microstructures are often produced by the recrystallization of a cold worked material, the smallest grain sizes (*ca.* 10 μm) being achieved in alloys containing large second-phase particles that promote recrystallization during the annealing of highly strained material. However, recent research has shown that the grain refinement limits imposed by conventional thermomechanical processing can be overcome by the application of very large plastic strains, and that sub-micron grain structures may be produced in many metallic materials. Such processing routes provide exciting new opportunities for the development of high-performance structural metallic materials.

A deformed metal has a large stored energy and, on annealing at an elevated temperature, will normally revert to a lower energy by the formation of new defect-free grains that grow to consume the deformed microstructure. This is a process of *discontinuous recrystallization*, and it is usually found that with increasing plastic deformation, the microstructural instability increases and the metal recrystallizes more readily (Humphreys & Hatherly 1995). However, it has been shown that sub-micron grain structures may be formed directly by very large strain deformation, with little or no subsequent annealing, and the formation and stability of such microstructures are discussed in the following sections.

2. Methods for large strain processing

Many industrially important metal-forming methods, such as rolling and extrusion, impart large plastic strains and, as will be discussed later, very fine grained microstructures may, in certain cases, be formed. However, during such processing, one or more dimensions of the work piece is continuously reduced and, eventually, foil or filamentary materials, having limited use for structural applications, are produced. There are a number of high-strain processing methods in which a sample can be deformed without any net change in its dimensions, and there is no limit to the strain that can be achieved, provided the material has sufficient ductility. The concept of using such redundant shape change processes to achieve ultra-high strain deformations and produce microstructural improvements in alloys is by no means new, and the method of forging and folding used to produce the Samurai sword is one such process. Two types of method are now commonly used to produce such large strains, those involving a redundant net strain, based on reversing the strain path for each cycle, and those where the sense of the deformation is always in one direction, but no shape change of the work piece results.

(*a*) *Redundant strain processes*

Redundant strain processes (figure 1) are probably the easiest to apply in commercial practice because they are cyclic in nature, and methods such as reciprocating extrusion have the added advantage that the sample is fully constrained, thereby allowing processing of less ductile materials. This method has been used to achieve redundant strains as high as 90 and grain sizes of the order of 2 μm have been achieved in pure aluminium (Richert & Richert 1986). The disadvantage of using a fully reversible strain path is that this is a less efficient way of storing dislocations in the material and of breaking up the original grain structure than a directional process, because the deformation mechanisms of the material are, to a surprisingly large extent, reversible. For example, it has been shown that the original grain shapes are largely restored on reversing large torsional deformations (Farag *et al.* 1968), and

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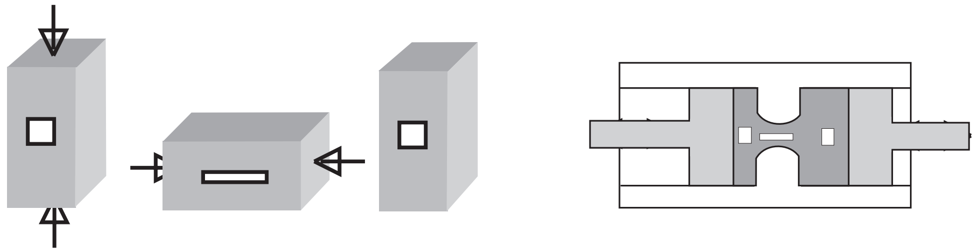

Figure 1. Examples of two techniques used to achieve high levels of redundant strain.

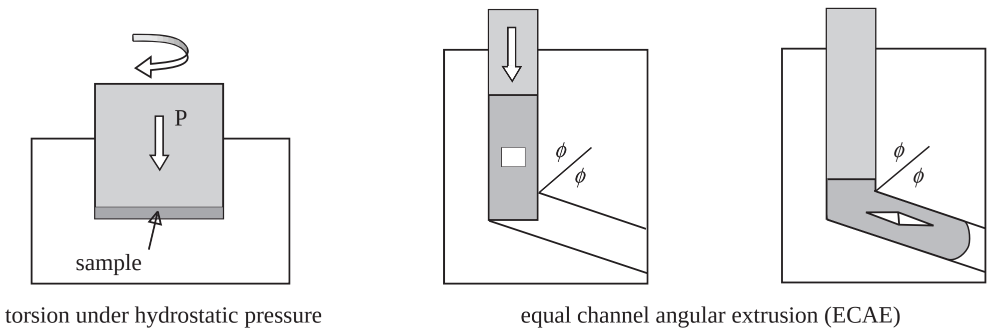

Figure 2. Examples of directional strain processes used to achieve very high plastic strains.

that the recrystallization kinetics is slower after reversing the strain path in multiple forging, compared to monotonic deformation to the same equivalent strain (Embury *et al*. 1992). However, in processes such as reciprocating extrusion that involve very complex strain patterns, the strain path is unlikely to be fully reversible.

(*b*) *Directional strain processes*

In directional strain processes the strain path is not reversed, although the deformation, which is usually predominantly by shear, can still be constrained within the shape of the work piece, and much larger strains than can be achieved by conventional processing are then possible. One method that has been used with a wide range of materials is that of torsion under hydrostatic pressure, adapted from the Bridgeman anvil (Bridgeman 1952; Saunders & Nutting 1984; Valiev *et al*. 1992). In this technique (figure 2), a thin disc is deformed in torsion using the friction provided by the application of a large hydrostatic pressure (*ca.* 5 GPa). The equivalent strains that have been induced with this method are typically of the order of 7, and, as discussed in § 3, grain sizes as fine as 0.2 μm have been produced by deformation at room temperature. This technique cannot readily be scaled up and is, therefore, most suitable for small-scale laboratory investigations.

(*c*) *Equal channel angular extrusion*

An alternative method is that of equal channel angular extrusion (ECAE) developed in the former Soviet Union by Segal and co-workers (Segal *et al*. 1981; Segal

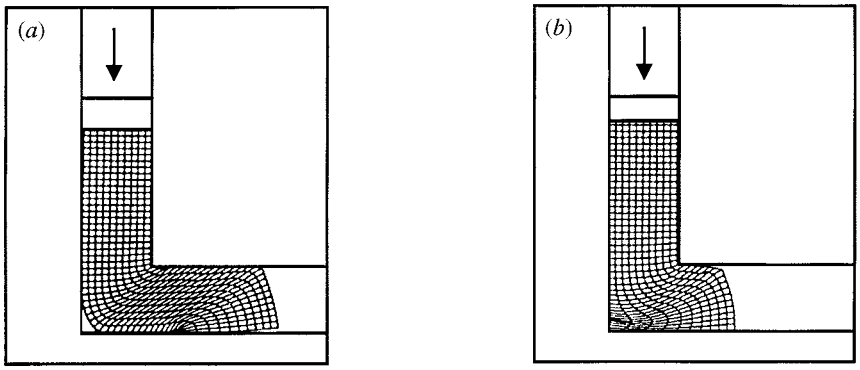

Figure 3. Finite-element model of the deformation of a sample during ECAE extrusion with a 90° die angle, without (*a*), and with (*b*) friction (Prangnell *et al*. 1997).

1995), which has been used to obtain much of the experimental data presented in this paper. Of the possible methods for obtaining very high plastic strains, it has arguably the simplest deformation mode, which under ideal conditions is a simple homogenous in-plane shear. This makes possible the quantitative interpretation of the development of the deformation microstructures. The technique can also be easily scaled up to produce substantial samples for property determination.

During ECAE, the sample is extruded in a closed die that has two intersecting channels of equal size (figure 2) offset at an angle 2ϕ. Assuming there is no friction and a sharp die corner, the sample will be subjected to a homogenous shear, apart from at its ends, which can be shown to be simply dependent on the die angle (2ϕ) by

$$\gamma = 2\cot\phi. \tag{2.1}$$

For multiple passes (n), the strains can be summed, and an equivalent strain (ε_{E}), which enables comparison with other deformation processes, can be defined (Segal *et al*. 1981) as

$$\varepsilon_{\mathrm{E}} = (2n/\sqrt{3})\cot\phi. \tag{2.2}$$

The above analysis can also be used to predict, in a material of grain size D_0, the separation of the original grain boundaries (D), as a function of strain, for a constant strain path:

$$D = D_0/(\sqrt{1+\gamma^2}). \tag{2.3}$$

Although the concept of an equivalent strain is useful for comparing different processes such as rolling and ECAE, the differences in the deformation modes between these processes lead to quite different slip activities, and the microstructures and textures developed will, therefore, differ. During ECAE, the grains are elongated by shear, and the separation of the original grain boundaries (D), as given by equation (2.3), reduces with increasing strain at a lower rate than for deformation by rolling (see § 3).

(i) *Modelling ECAE deformation*

There are a number of factors, including friction and die shape, that make ECAE deformation more complicated than indicated in the above analysis. Recently, the

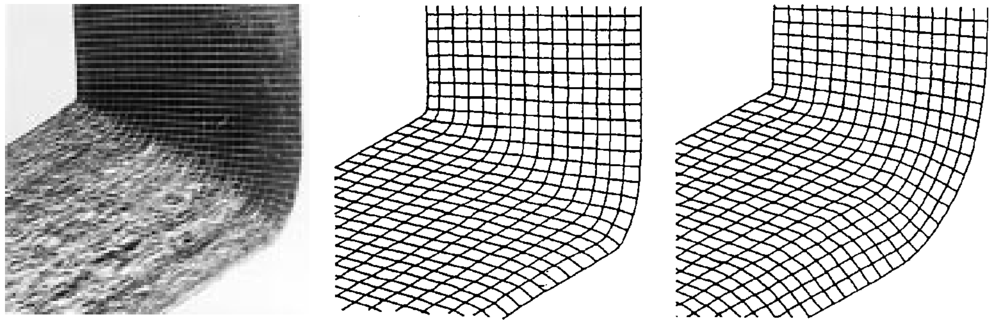

Figure 4. Experimental measurement of ECAE deformation using a split sample. (*a*) Micrograph of a scribed sample deformed under high-friction conditions. (*b*) A tracing of the grid shown in (*a*). (*c*) A tracing of a grid on a specimen deformed under low-friction conditions (Gholinia *et al*. 1998).

deformation behaviour in the die has been modelled numerically (Prangnell *et al*. 1997; Bowen *et al*. 1999) and studied using a Plasticine analogue (Wu & Baker 1997). The early stages of metal flow are complex, as is shown in figure 3, and, on initial deformation, the elements at the base of the billet are compressed. The material on the bottom right-hand corner, that is unconstrained by the second channel opening, is only slightly deformed and is mainly rotated, spilling over the material on the base of the billet, because it is pushed by the sheared material following behind. However, a substantial portion of the billet appears to be uniformly deformed, once its end has passed through the die corner. It is, thus, advisable to ignore the ends when analysing the samples. Finite-element (FE) modelling shows that a die angle $2\phi = 90°$ (used by many workers) and high friction, can lead to better filling of the die corner but a larger zone at the end of the sample, where the deformation is initially inhomogeneous (figure 3). With a shallower die angle and lower friction, the sample end slides sideways more readily, which reduces the die corner filling and the size of the end zone.

The effects of friction and the application of a back pressure on the homogeneity of shear have been investigated by Bowen *et al*. (1999), who compared experimentally deformed split samples with a scribed grid on the centre plane with the predictions of FE modelling. The deformed grids shown in figure 4 were in good agreement with the FE simulations, and both demonstrated that the most uniform deformation across the sample occurs when a back pressure is applied and the die is fully filled (figure 4*a*, *b*). In this case, the shear strain measured is close to that predicted by equation (2.1), which can be considered as an upper bound for an idealized homogeneous shear. When the sample does not fully fill the die (figure 4*c*), or if a radiused internal die corner is used, the sample is bent rather than sheared (Segal *et al*. 1981), resulting in the outer surface being stretched in tension, through an arc, and then compressed as it moves away from the deformation zone. Under such conditions, the outer surface of the sample is subjected to a redundant tensile strain rather than a shear, and it is this that probably explains the observations of Nakashima *et al*. (1998) that a radiused die is less effective than a sharp die in forming an ultra-fine grain structure. The deformation pattern will also be dependent on the die angle (ϕ)

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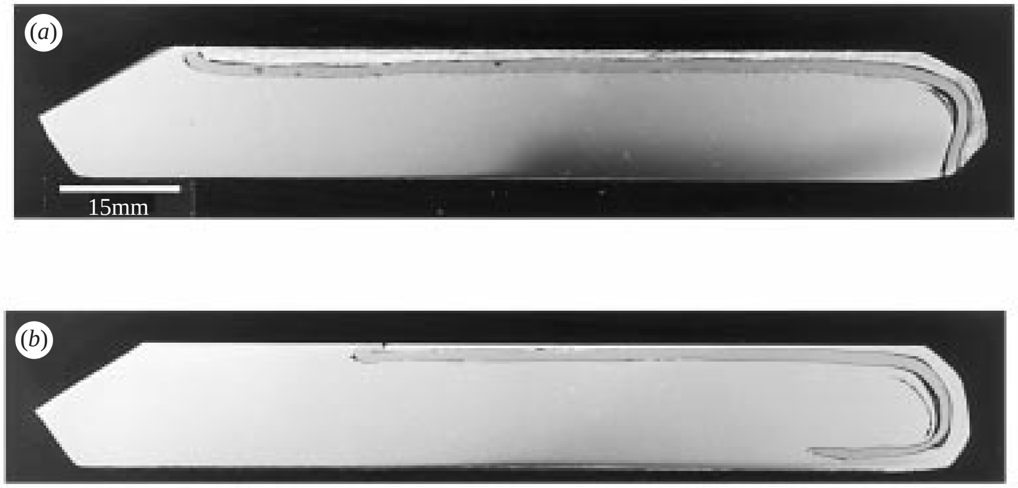

Figure 5. Cross-section of an aluminium specimen containing a copper wire, which has been deformed by ECAE ($2\phi = 135°$) for (*a*) two passes; (*b*) five passes.

(Nakashima *et al*. 1998; Prangnell *et al*. 1997) and, in particular, when $2\phi \sim 90°$, it is unlikely that equations (2.1)–(2.3) will be strictly valid.

(ii) *Secondary deformations*

It is important to realize that although the sample appears largely to maintain its dimensions during processing (see figures 2 and 3), the effect of a repeated simple shear is to increase the length. This implies that secondary deformations are required to maintain the shape of the billet (Gholinia *et al*. 1998). The inconsistency between the sample retaining its approximate dimensions after repeated passes, and the continuously increasing shear strain with each pass, has been investigated by Bowen *et al*. (1999) using a sample containing an embedded copper wire. In figure 5, it can be seen that the change in sample length is accommodated by the sample being wrapped back on itself. Material starting on the sample top is sheared and then rotated around at the sample end, ending up on the bottom. This occurs in increments corresponding to each pass, and the resulting facets can be seen on the end of the sample. These results are in agreement with those of the Plasticine simulations of Wu & Baker (1997). After a large number of passes, a substantial proportion of the sample will have rotated around each end of the billet. Compared to the massive shear strains developed after multiple passes, the secondary deformations are relatively small, although their effects on the break-up of the initial grain structure and on texture development have not been determined yet.

(iii) *Sample orientation*

In ECAE it is not necessary to keep the work piece orientation the same for each repeated pass. Several permutations are possible. The sample can be rotated end over end, reversing the strain path between each pass, or it can be rotated around its axis. As has been pointed out previously, reversing the strain path is not desirable because this is a less efficient means of encouraging the break-up of the initial grain

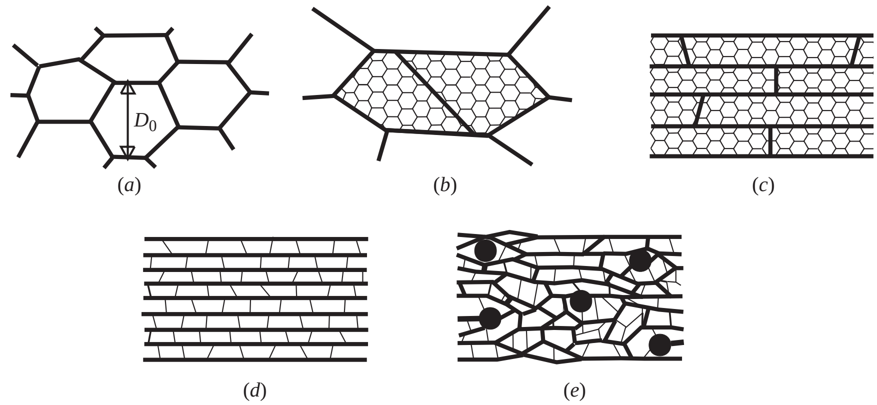

Figure 6. Schematic diagram showing the development of microstructure with increasing strain. (*a*) Initial grain structure; (*b*) subgrains and grain subdivision; (*c*) alignment of HAGBs; (*d*) ribbon grain structure; (*e*) break-up of ribbon grains by large second-phase particles.

structure. Iwahashi *et al*. (1998) suggested that the most effective way of breaking up the grain structure during ECAE is to rotate the sample 90° around its axis between each pass. This procedure leads to a more complex strain path than if the orientation of the sample remains constant, and may destabilize the distorted grains, leading to the formation of more high-angle grain boundaries (HAGBs).

3. Microstructural development during large strain deformation

(*a*) *General features*

If a metal is plastically deformed at ambient temperatures, then significant changes occur in the microstructure, in particular to the HAGBs ($\theta > 15°$), *low-angle grain boundaries* (LAGBs) ($\theta < 15°$), and the density and distribution of *dislocations*.

Careful research during the past decade has revealed many details of the development of microstructure during the low-temperature deformation of metals and these are reviewed elsewhere (Hansen & Juul Jensen, this issue; Gil Sevillano, this issue). In this paper, we focus on those aspects that are most relevant to the formation of fine-grain microstructures. Although deformation occurs by the movement of dislocations, at large strains in materials of high-stacking fault energy, such as aluminium alloys, most of the dislocations are assimilated by dynamic recovery into LAGBs, which form a three-dimensional network of subgrains within the grains, and relatively few free dislocations are found in the subgrain interiors (Lloyd & Kenny 1980). To a first approximation, therefore, the microstructure of a highly deformed metal may be described in terms of the distribution of the grain and the subgrain boundaries.

Figure 6 is a schematic diagram of the development of microstructure during the cold rolling or plane-strain compression of a polycrystal that has an initial grain size (D_0). At intermediate strains (figure 6*b*), we note three important microstructural developments, as follows.

(1) The original grains are deformed to a shape determined by the applied strain and strain path, and the area of the HAGB is increased.

(2) A cell or subgrain structure has developed within the grains. With increasing strain, the cell size decreases and the misorientation between cells increases until both tend to a steady state at strains of *ca.* 1–2 (see, for example, Humphreys & Hatherly 1995). Typically, the subgrain size is *ca.* 0.2–0.5 μm, and the misorientation *ca.* 1–5°. It is thought that such steady-state subgrain structures are the result of the subgrain boundaries being constantly altered, formed and removed by the passage of dislocations during deformation, and they have been termed *incidental dislocation boundaries* (Kuhlmann-Wilsdorf & Hansen 1991).

(3) Some medium- or high-angle boundaries may be created within the old grains by grain subdivision. Such boundaries may separate regions within a grain that are deforming on different sets of slip systems, such that their orientations diverge with increasing strain until stable end-orientations are reached. High-angle boundaries may also be formed as the result of coarse slip associated with shear banding. Several different types of such boundary, often classified as *geometrically necessary boundaries* (Kuhlmann-Wilsdorf & Hansen 1991), have been identified.

At large strains (figure 6*c*), the distinction between the original and the new high-angle boundaries is lost, and the high-angle boundaries tend to be aligned in the rolling plane. Eventually, at very large strains, the spacing between the high-angle boundaries will be reduced to the subgrain size (figure 6*d*).

(*b*) *High-angle grain boundaries*

The increase in the area of the HAGB due, simply, to the shape change of the original grains is readily calculated, and depends both on the amount of strain and the nature of the deformation (Gil Sevillano *et al*. 1980). For example, in the case of plane-strain compression (rolling), the relationship between the original grain size (D_0), the high-angle boundary spacing at large strains, perpendicular to the rolling plane (D), and the true strain (ε_{T}), is given by

$$D = D_0 \mathrm{e}^{-\varepsilon_{\mathrm{T}}}, \tag{3.1}$$

which may be compared with equation (2.3), which is the equivalent relationship for ECAE.

(*c*) *Grain subdivision*

The formation of medium- to high-angle boundaries within existing grains has been known for many years (see, for example, Boas & Hargreaves 1948). However, it is only recently that this phenomenon has been extensively studied and quantified (see, for example, Hughes & Hansen 1997; Hansen & Juul Jensen, this issue). Of particular relevance to the present discussion is the extent of such grain subdivision. It is known that this is affected by the material (Hughes & Hansen 1997), the grain orientation (Lee *et al*. 1994), the orientation relationships of adjacent grains (Hutchinson 1974), the strain and deformation mode, and the initial grain size and shape.

The effect of strain on grain subdivision in commercial-purity aluminium (AA1050) deformed by ECAE at room temperature in a die with $2\phi = 135°$ is shown in figure 7, where the measured high-angle boundary separation (D) divided by the theoretical

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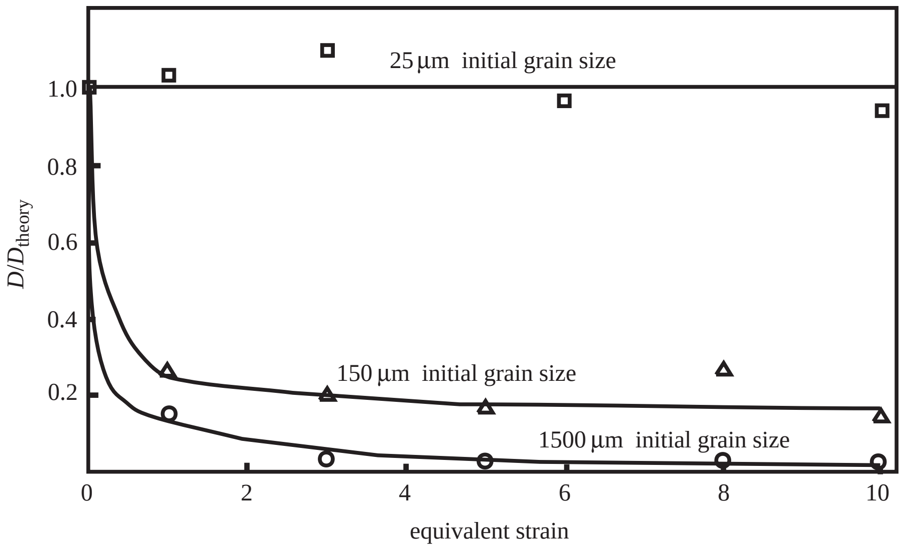

Figure 7. The effect of strain and initial grain size on grain subdivision in aluminium AA1050. The samples were given a recovery anneal at 200 °C before examination, in order to sharpen the deformation structure before EBSD.

boundary spacing that would result if there were no grain subdivision (D_{theory}) is plotted as a function of strain. It is seen that grain subdivision is more extensive for the coarse-grained material at low strains, but that at larger strains, there is little further subdivision, and it is also seen that there is little evidence of grain subdivision for finer-grained material.

The effect of the initial grain size on grain subdivision in aluminium is shown in figure 8, which includes data from a number of investigations, and the reduction in grain subdivision for small grain sizes is evident. Lee *et al*. (1994) have modelled grain subdivision and suggest that the number of subdivisions (D_{theory}/D) should be proportional to the square root of the initial grain size, and the data for larger grain sizes are broadly consistent with this. However, for grain sizes below *ca.* 25 μm, it is clear that grain subdivision does not play an important role in development of the deformation microstructure in aluminium.

(*d*) *The role of deformation heterogeneity*

It is evident from the preceding discussion that, for large strains, a large area of the HAGB is generated in fine-grained metals purely by enlargement of the existing boundaries. If the grains were to deform in the ideal manner shown in figure 6*d*, then the resulting microstructure would contain very thin ribbon grains and could not be accurately described as a 'fine-grained microstructure'. It is well established that very large torsional or rolling strains at elevated temperatures may produce very long, thin, ribbon grains (Jonas *et al*. 1969; Gholinia *et al*. 1997), although the limits to such a process, at which the onset of fragmentation of the ribbons occurs, have not been established. There are several factors that encourage the breakdown of such planar boundary structures.

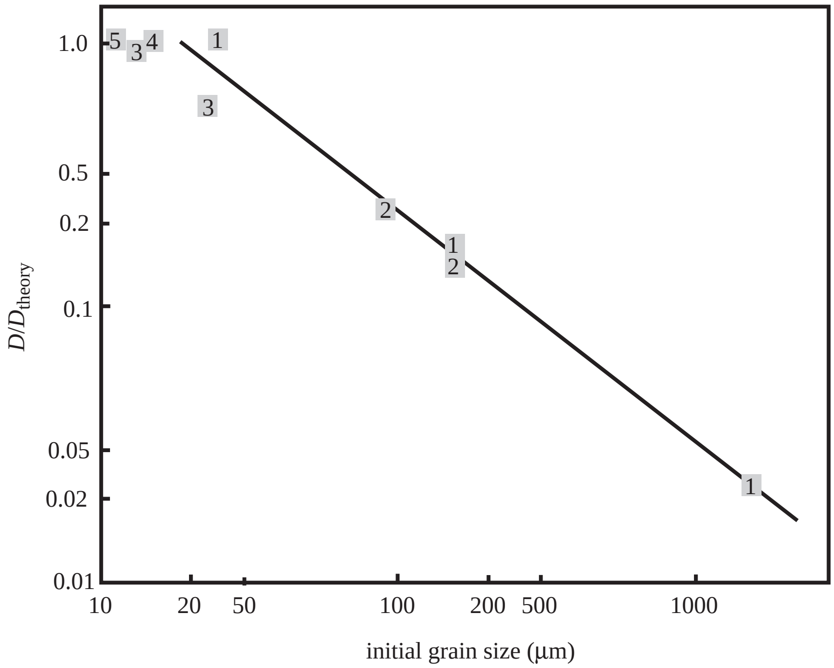

Figure 8. The effect of initial grain size on grain subdivision for several aluminium alloys deformed to strains greater than 1. 1: AA1050, this investigation. 2: Hughes & Hansen (1997). 3: Saeter & Nes (1996). 4: AA8014, this investigation. 5: Oscarsson *et al*. (1992).

(a) *Large non-deformable second-phase particles* cause inhomogeneous deformation of the adjacent metal that will lead to strain gradients and the formation of additional high-angle boundaries, which will tend to destroy the planarity of the boundaries (Humphreys & Hatherly 1995).

(b) *Complex deformation paths* such as are achieved with some of the redundant deformation methods discussed in § 2, e.g. reciprocating extrusion or ECAE with the sample rotated between passes, generate high-angle boundary area but do not produce aligned planar boundaries.

(c) *Shear banding* commonly occurs in heavily rolled metals (see Humphreys & Hatherly 1995). Bands of intense shear, independent of the grain structure, often occur at *ca.* 35° to the rolling plane, and may cause significant displacements of the grain structure. The tendency for shear banding is increased by a low work hardening rate and may be affected by precipitates or solutes (Hutchinson, this issue).

(d) *Grain-scale deformation.* The individual grains in a sample will have different flow stresses, and those of high Taylor factor will be more resistant to deformation. This effect, which may be significant in steels (Hutchinson, this issue), leads to local strain variations that reduce the planarity of the boundaries.

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(*e*) *The role of deformation temperature*

During elevated temperature deformation, grain boundaries become serrated by migration due to the tension of interconnecting low-angle boundaries. The extent of these serrations is approximately equal to the subgrain size and, thus, when the high-angle boundary spacing is reduced by straining, to the order of the subgrain diameter, grain impingement occurs (Humphreys 1982). This phenomenon, which is usually termed *geometric dynamic recrystallization* (McQueen *et al*. 1985), is, therefore, closely related to the geometric increase in high-angle boundary area found during low-temperature deformation. A further factor that aids the formation of a fine grain structure during deformation at elevated temperatures is the onset of *grain boundary sliding*. It is likely that this process can occur once a critical amount of HAGB is present (Cullen *et al*. 1994), and once operative it will lead to further HAGB formation and the formation of a more equiaxed grain structure. During high-temperature deformation, there are, however, also factors that lower the rates of formation of high-angle boundary and favour the formation of the ribbon grains discussed above. For example, grain subdivision is less extensive and the effect of large second-phase particles on deformation heterogeneity is diminished (see Humphreys & Hatherly 1995).

(*f*) *Microstructures of aluminium alloys after large strain deformation*

The microstructures of Al–3%Mg alloys deformed in high-pressure torsion to strains of about 7 at room temperature have been reported by Horita and co-workers (see, for example, Horita *et al*. 1997; Wang *et al*. 1996). Cellular structures of dimension *ca.* 0.1–0.2 μm containing free dislocations are observed in the transmission electron microscope (TEM), and, although detailed analysis of the boundary misorientations has not been made, the extensive arcing of diffraction patterns has been interpreted as being due to a large number of HAGBs. High-resolution TEM imaging shows that many of the boundaries are highly faceted and contain defects, indicating that they are in a high-energy non-equilibrium configuration. The microstructures evolved during ECAE at room temperature of a number of other aluminium alloys have been reported by Horita and co-workers (see, for example, Horita *et al*. 1998), and equiaxed grain structures of *ca.* 0.5–1 μm are reported. The effects of die angle and sample rotation on microstructures in 99.99% aluminium have been investigated by Nakashima *et al*. (1998), who found that equiaxed 'grains' of *ca.* 1 μm are developed most efficiently ($\varepsilon_{\mathrm{E}} \sim 4$) in a die with $2\phi = 90°$ if the specimen is rotated by 90° between passes.

A number of aluminium alloys have been deformed to large strains by the Manchester group, either by ECAE or by rolling, and detailed characterization of the resulting boundary misorientations has been carried out by TEM imaging and high-resolution SEM and EBSD. There are a number of similarities between the microstructures of these materials, as summarized in table 1. In all cases, after large strain deformation, well-defined grain/subgrain structures with few free dislocations are observed (figure 9*a*). The grains tend to be elongated in the direction of rolling or shear with an aspect ratio of 1.5–3, and the separation of the high-angle boundaries parallel and perpendicular to the rolling/shear plane, as shown in table 1, is typically less than 1 μm.

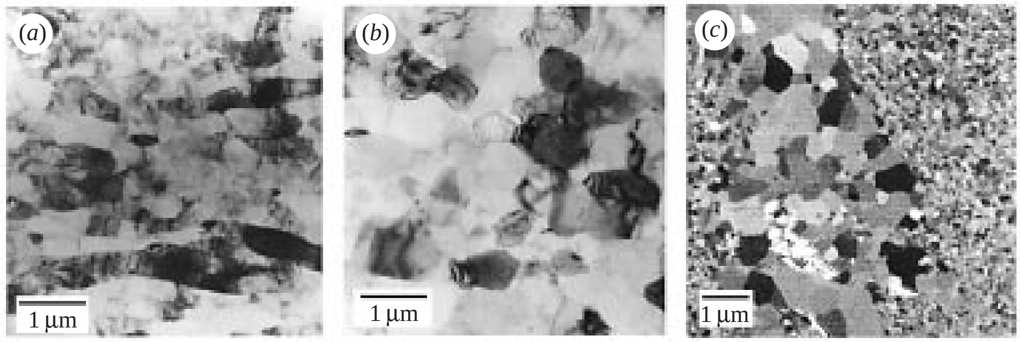

Figure 9. TEM images of an Al–3%Mg–0.2%Zr alloy deformed by ECAE at 200 °C to an equivalent strain of 9.8. (*a*) Elongated grain structure in the deformed alloy. (*b*) Equiaxed microstructure formed on annealing at 300 °C. (*c*) Abnormal grain growth during annealing at 300 °C. In all cases, the direction of shear is horizontal (Gholinia *et al*. 1998).

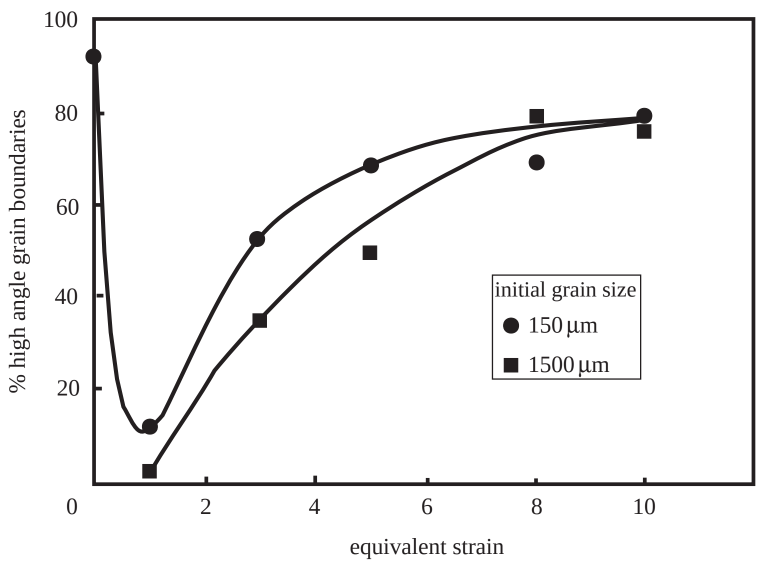

Figure 10. The percentage of HAGBs as a function of strain in AA1050 deformed by ECAE. The samples were given a recovery anneal at 200 °C before examination (Harris *et al*. 1998).

The relative number of high- and low-angle grain boundaries varies with strain in a complex manner, as shown in figure 10. The theoretical fraction of high-angle boundaries in a random grain assembly is *ca.* 0.97, but, at small strains, low-angle boundaries are formed and the fraction of HAGB decreases. However, as discussed in § 3, the high-angle boundary area increases continually with strain, whereas the subgrain size reaches a steady state of *ca.* 0.5 μm at large strains. Therefore, the fraction of HAGB increases at larger strains, tending to saturate at *ca.* 0.65–0.75 (table 1). This value is often smaller than that of the undeformed material, because transient low-angle boundaries are continually being created during deformation and also, the development of a strong deformation texture ensures a large number of 'crystallographically necessary' medium-/low-angle boundaries in the microstructure.

Table 1. *Large strain deformation microstructures*

aluminium alloy	deformation: method	deformation: temperature	deformation: true or equivalent strain	HAGB spacing (μm): grain length	HAGB spacing (μm): grain thickness	HAGB fraction
AA1050	ECAE ($2\phi = 135°$)	RT	10	2.3	0.9	0.65
Al–3%Mg–0.2%Zr	ECAE ($2\phi = 120°$)	200 °C	9.8	0.45	0.35	0.77
AA8014	rolling	RT	3.5	0.54	0.37	0.69

It should be emphasized that although these highly deformed materials contain a very large fraction of high-angle boundaries, which approaches that of a highly textured recrystallized alloy, they cannot be considered to be 'recrystallized grain structures'. The microstructures are those predicted to be formed by the normal processes of deformation and dynamic recovery, and they are similar to those shown schematically in figure 6*e*.

4. Annealing of highly deformed alloys

(*a*) *Low-temperature annealing*

Annealing of the heavily deformed specimens in table 1 in the temperature range 150–250 °C results in small microstructural changes. In all cases, the grain structures become slightly coarser and more equiaxed, as shown in figure 9*b*, and there is a small increase in the fraction of HAGB. The materials retained their strong deformation textures with only minor changes in the proportions of the major components. This low-temperature annealing process, which transforms the heavily deformed microstructure to an equiaxed grain structure by minor boundary rearrangements, has been observed in several Al–Fe–Si alloys (see, for example, Oscarsson *et al*. 1992; Davies *et al*. 1997; Horita *et al*. 1998), and can be described as one of continuous recrystallization.

(*b*) *Annealing at higher temperatures*

At higher temperatures, the annealing behaviour of the heavily deformed alloys of table 1 is significantly different from that of similar material deformed to lower strains. Whereas a moderately deformed alloy will recrystallize discontinuously at *ca.* 300 °C, as shown in figure 11*a*, the heavily deformed alloys retain the fine-grain structure formed by continuous recrystallization, as shown in figure 11*b*. Similar effects have been reported by other authors (Oscarsson *et al*. 1992; Davies *et al*. 1997; Harris *et al*. 1997), and for these alloys, there is, therefore, a critical deformation, below which recrystallization is discontinuous, and above which it is continuous.

It is, therefore, established that large strains may lead to fine-grained microstructures that are resistant to discontinuous recrystallization, and Oscarsson *et al*. (1992)

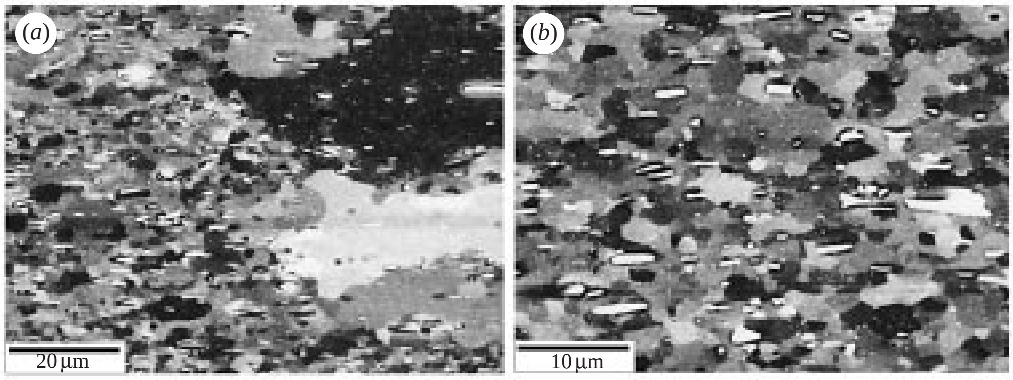

Figure 11. Annealing of cold-rolled AA8014 at 300 °C. (*a*) 90% cold-rolled specimen is partly recrystallized. (*b*) 98% cold-rolled specimen does not recrystallize.

were the first to correctly identify such stability as being due to the large fraction of HAGB formed during deformation. A theoretical analysis of the stability of cellular microstructures, based on the energy and mobility of boundaries, has recently been given by Humphreys (1997*a*), who showed that low-angle grain boundary microstructures ($\theta < 10°$) are inherently unstable and that if recrystallization does not intervene, then discontinuous subgrain growth is predicted. However, if the mean misorientation (θ) between (sub)grains is larger, discontinuous growth within the microstructure becomes less extensive, and for $\theta > 10°$, a microstructure that is stable against discontinuous growth results, and only normal grain growth is possible. For a mean subgrain misorientation of 3°, it is estimated that the critical fraction of HAGB is 0.70–0.75, which is in agreement with the computer simulations of Oscarsson *et al*. (1994) and consistent with the behaviour of the materials of table 1.

(*c*) *Grain growth*

The fine-grained microstructures formed by large strain deformation and low-temperature annealing have a large energy associated with the grain boundaries, and, at higher temperatures, further microstructural changes will occur to reduce the boundary area. The grain growth behaviour is found to depend on the distribution of small second-phase particles in the alloy, and the ratio of volume fraction (F_v) to particle diameter (d) is critical. Three cases can be distinguished, as follows.

(I) *No small particles* ($F_v/d \sim 0$). Alloys that are single-phase or that contain only large second-phase particles (e.g. Al–Fe–Si alloys such as AA1050 and AA8079) undergo normal (continuous) grain growth at elevated temperatures (Oscarsson *et al*. 1992; Harris *et al*. 1997; Davies *et al*. 1997; Horita *et al*. 1998).

(II) *Some small particles (medium* F_v/d*)*. The AA8014 and Al–Mg–Zr alloys of table 1 undergo some normal grain growth; however, at temperatures above *ca.* 300 °C, abnormal or discontinuous grain growth occurs, as is shown in figure 9*c* (Gholinia *et al*. 1998).

(III) *Many small particles (large* F_v/d*)*. In age hardening aluminium alloys AA7075 and AA2024 (Horita *et al*. 1998) and in spray-cast dispersion hardened mate-

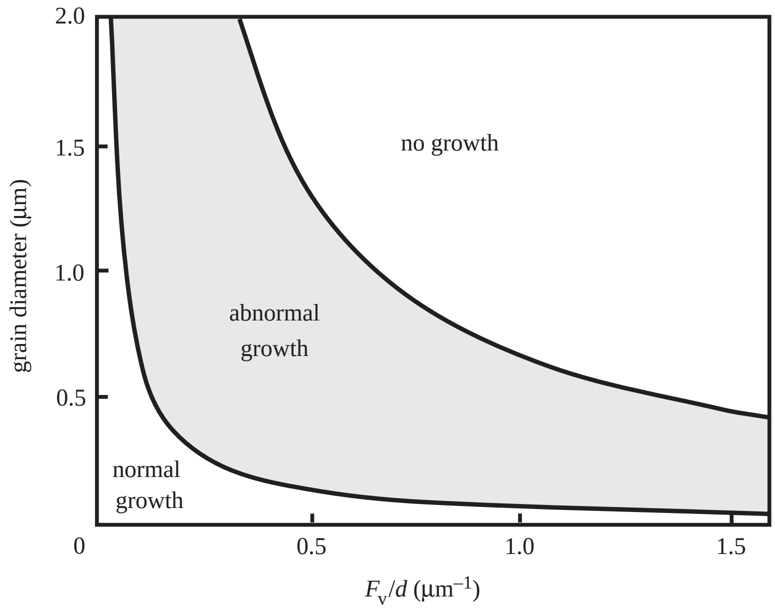

Figure 12. Regimes of normal and abnormal grain growth for particle-containing alloys (after Humphreys 1997*a*, *b*).

rials (Gholinia *et al*. 1997), there is little evidence of normal or abnormal grain growth, and the ultra-fine grain structure is retained to higher temperatures, subject to the particle stability.

The theory of the stability of cellular microstructures has been extended to two-phase alloys (Humphreys 1997*b*) and may be used to explain the effect of particles on the normal and abnormal grain growth of fine-grain microstructures. Figure 12 illustrates the influence of the pinning particles (F_v/d) and the grain size (D) on the stability of fine-grain structures. It should be noted that in constructing this figure, the effects of texture have been ignored. The figure explains the three types of grain growth behaviour discussed above. If there are few second-phase particles (e.g. AA1050) (case I), then it is seen that normal grain growth is predicted to occur during high-temperature annealing. However, as F_v/d increases (case II), abnormal grain growth is increasingly likely for smaller grain sizes. In an alloy such as the AA8014 material of table 1, which has $F_v/d \sim 0.1\ \mu m^{-1}$, the 0.5 μm diameter grains are predicted to undergo abnormal grain growth after a small amount of normal grain growth, as is found. In order to maintain a very small grain structure at high temperatures (case III), sufficient particles must be present to prevent abnormal grain growth, and this requires $F_v/d > 1.5\ \mu m^{-1}$ for 0.5 μm diameter grains.

5. Summary

From the preceding discussion, we can summarize the factors that are necessary for the production of a sub-micron grain structure in aluminium alloys by large strain processing, and which will influence its stability at elevated temperatures.

(a) Formation of a sub-micron grain structure

The main criterion is the generation of sufficient HAGB area, such that the fraction of HAGB is greater than 0.65–0.75. Factors that promote this condition are a small initial grain size, a large strain, and a deformation process with a complex deformation path that prevents formation of planar boundary arrays. Dynamic recovery processes are important in forming the fine grain/subgrain structure, and factors that impede dynamic recovery, such as high solute content (e.g. Mg in Al), and closely spaced small second-phase particles are likely to be detrimental.

A low deformation temperature may be beneficial as there is more grain subdivision and plastic heterogeneity will be introduced by any large (greater than 1 μm) second-phase particles. Although the latter effect is lost during high-temperature deformation, boundary serration will increase the amount of HAGB. However, this effect will be diminished if there are sufficient small particles to pin the high-angle boundaries.

(b) The stability of a sub-micron grain structure at high temperatures

Although it may be sufficient to produce a fine grain structure, which is stable at temperatures of up to 150–200 °C, there may be a requirement to maintain such a microstructure to higher service temperatures. The energy stored in a sub-micron grained material is considerable, and stability can only be achieved if both normal and abnormal grain growth can be suppressed by the presence of a high density of small second-phase particles. Such particles may be present during the deformation processing, although, as discussed above, they will generally hinder the formation of the fine-grained structure. Alternatively, the material may be deformed in a solution-treated condition and the particles introduced during a subsequent ageing heat treatment. The maximum temperature to which any fine-grained microstructure can be maintained is, of course, dependent on the thermal stability of the second-phase particles.

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Discussion

B. Hutchinson (*Swedish Institute for Metals Research, Stockholm, Sweden*). In Professor Humphreys's micrograph of abnormal grain growth, the large grains appeared to occur in colonies. Is this a characteristic behaviour, and if so, what is its origin?

F. J. Humphreys. In most of our materials which show abnormal grain growth, the abnormally growing grains are distributed reasonably randomly. Professor Hutchinson is correct in pointing out that the micrograph of figure 9*c* does show abnormal growth only in certain regions of the sample, and the unusual behaviour of this Al–Mg–Zr alloy is thought to be associated with non-uniform distribution of second-phase particles in the original casting.

L. M. Brown (*Cavendish Laboratories, University of Cambridge, UK*). Would it be true that in these processes in making use of a large amount of redundant deformation some hydrostatic pressure is required to prevent cavitation at the particles?

F. J. Humphreys. The amount of cavitation occurring during deformation of most aluminium alloys to large strains is generally very small because of the strong interfacial bonding, and cavitation is not a problem when such alloys (e.g. AA8014) are rolled to very large strains at room temperature. During equal channel angular extrusion, friction between the specimen and die results in significant hydrostatic pressure.

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Although our ECAE rig does have a capability for imposing a back pressure on the sample, we use this only to ensure that the sample conforms to the die shape (figure 4), and have not found significant cavitation in our samples, even when no back pressure is applied.

M. J. Stowell (*Saffron Walden, Essex, UK*). Could Professor Humphreys comment on the potential for scaling up the ECAE process?

F. J. Humphreys. In principle, there are no major problems in scaling up the ECAE process apart from the consequent increases in load on both the die and the press, and Russian researchers have apparently extruded bars of diameters greater than 50 mm. We are currently designing dies capable of extruding 45 mm diameter high-strength aluminium alloys.

J. Gil Sevillano (*CEIT, University of Navarra, San Sebastián, Spain*). Because of the sharp kinking in the die that is repeated always in the same sense, important internal microstresses are probably being developed. Has Professor Humphreys measured them or remarked on some of their effects? (They could be important for technical applications.)

F. J. Humphreys. We have not yet investigated the magnitude of any internal stresses in alloys deformed by equal channel angular extrusion. However, as the higher-strength materials are generally deformed at elevated temperatures, it is likely that recovery processes will eliminate most of the internal stress.

P. J. Withers (*Manchester Materials Science Centre, UK*). What are the implications for joining such materials?

F. J. Humphreys. Although this area has not yet been investigated, it is clear that materials with such fine-grain structures will be intrinsically unstable, and this may cause problems in joining. If the amount of boundary pinning from second-phase particles is small (figure 12), then any heating is liable to result in abnormal grain growth. However, if there are sufficient particles to prevent abnormal grain growth, then processes such as friction or spot welding or diffusion bonding may be successful.

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